# The Anisotropy of the London Penetration Depth and the Upper Critical Field in C-doped MgB<sub>2</sub> Single Crystals from Reversible Magnetization

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#### Abstract

We have studied the anisotropy of the London penetration depth of carbon doped MgB<sub>2</sub> single crystals, which was obtained from reversible magnetization measurements with the magnetic field both parallel and perpendicular to the c-axis. Similar to the pure MgB<sub>2</sub>, the anisotropy of the upper critical field  $\gamma_{H}$  decrease with temperature while the anisotropy of the London penetration depth  $\gamma_{\lambda}$  slowly increases with temperature. However, the temperature dependence of  $\gamma_{H}$  is drastically reduced and the value of  $\gamma_{\lambda}$  becomes nearly ~1 as C is introduced. These results indicate that C substitution increases impurity scattering mainly in the  $\sigma$  bands. The temperature dependence of the anisotropies agree well with the theoretical predictions with impurity scattering.

Keywords : C-doped MgB<sub>2</sub>, reversible magnetization, upper critical field, penetration depth

## 1. Introduction

It is now well established that  $MgB_2$  is a two-gap superconductor with two-band: quasi-twodimensional  $\sigma$  bands and three-dimensional  $\pi$  bands [1-3]. One of the consequences of the two-band nature is the strong temperature dependence of the anisotropy of the upper critical field  $(\gamma_H \equiv H_{c2}^{ab} / H_{c2}^{c})$  [4], which is in contrast to the single-gap Ginzburg-Landau theory. The strong temperature dependence of  $\gamma_{H}$  is thought to result from the interplay of two bands. Theoretical works have revealed that two different bands affect the behavior of  $\gamma_H$  such a way in which the anisotropic

 $\sigma$  bands dominate in low temperature region while at temperatures near  $T_c$ , contribution from the isotropic  $\pi$  bands increases [5-7].

Another consequence of the multi-band nature of MgB<sub>2</sub> is that the anisotropy of the penetration depth ( $\gamma_{\lambda}$ ), as well as the anisotropy of  $H_{c2}$ , may no longer be described by a single parameter [7-10]. In MgB<sub>2</sub>,  $\gamma_{\lambda}$  is not necessarily the same as  $\gamma_{H}$  since the penetration depth depends on the total number of charge carriers from both the  $\sigma$  band and the  $\pi$  band while  $H_{c2}$  is mainly determined by the  $\sigma$  band. According to Kogan's calculation based on weak-coupling theory [8],  $\gamma_{\lambda}$  is isotropic at low temperature and increases to 2.5 near  $T_{c}$ . That behavior was confirmed by the first-principle calculations for the electronic structure and the

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electron-phonon interaction [11], which showed that the effect of impurity scattering changed the exact value of  $\gamma_{\lambda}$ . The above anomalous behaviors of  $\gamma_{H}(T)$  and  $\gamma_{\lambda}(T)$  for MgB<sub>2</sub> single crystals were confirmed by magnetization measurements [12, 13].

When impurity scattering increases, the behavior of  $H_{c2}$  is modified. According to the dirty-limit two-gap theory, the shape of the  $H_{c2}(T)$  curve depends on the diffusivities of the  $\sigma$  and the  $\pi$ bands.  $H_{c2}(0)$  is determined by a minimum diffusivity(dirtier bands) while  $H_{c2}(T)$  is controlled by a maximum diffusivity (cleaner bands) at  $T \approx T_c$ . When the  $\sigma$  bands are dirtier, an upward curvature should appear near  $T_c$ , and  $\gamma_H$  should decrease with temperature. In contrast, when the  $\pi$  bands are dirtier, a huge increase in  $H_{c2}(T)$  should appear at low temperatures without an upward curvature near  $T_c$ , and  $\gamma_H$  should increase with temperature.

In this paper, we investigated the effect of C doping on the temperature dependence of the anisotropies of the penetration depth  $\gamma_{\lambda}$ , and the upper critical field  $\gamma_H$  of MgB<sub>2</sub> single crystals by measuring reversible magnetization for both  $H \parallel c$ and  $H \parallel ab$ . The reversible magnetization was analyzed by using the Hao-Clem model and the London model for  $H \parallel c$  and  $H \parallel ab$ , respectively.  $\gamma_{H}$  and  $\gamma_{\lambda}$  were found to show an opposite temperature dependence as shown in pure MgB<sub>2</sub>;  $\gamma_H$ decreases with temperature while  $\gamma_{\lambda}$  gradually increases with temperature. However, temperature dependence of  $\gamma_{H}$  is greatly reduced and  $\gamma_{\lambda}$ becomes more isotropic as C is introduced. These results agree well with the dirty-limit two-band theory.

#### 2. Experiments

 $Mg(B_{0.95}C_{0.05})_2$  single crystals were grown using a high pressure technique, which is explained in detail in previous reports [14, 15]. Concentration of C was determined by using both electron probe X-ray



Fig. 1. Temperature dependence of the reversible magnetization,  $4\pi M(T)$ , of C-doped MgB<sub>2</sub> single crystals for various field ranges of  $H \parallel c$  (data for only low fields are shown).

microanalysis and electron dispersive X-ray spectroscopy. 12 relatively hexagonal-shaped single crystals with typical dimensions of  $200 \times 100 \times 25 \,\mu\text{m}^3$  were collected on a substrate with their *c* axis aligned perpendicular to the substrate surface. The values of the transition temperature  $T_c$  and the transition width  $\Delta T_c$  determined from the low-field magnetization were 26 K and 2.4 K, respectively.

The measurement of the reversible and the irreversible magnetization were carried out by using a superconducting quantum interference device magnetometer (Quantum Design, MPMS-XL) with the fields up to 5 T applied parallel and perpendicular to the c axis of the sample.

## 3. Results and Discussion

Figure 1 shows, as an example, the temperature dependence of the reversible magnetization,  $4\pi M(T)$ , measured in the field range  $0.1 \text{ T} \le H \le 0.9 \text{ T}$  with  $H \parallel c$ . The reversible region was determined in the temperature ranges at which the criterion  $M_{FC}/M_{ZFC} \ge 0.95$  holds [16]. The reversible curves shifted to lower temperatures as the field was increased.

Fig. 2. Temperature dependence of the upper critical field for  $H \parallel c$  and  $H \parallel ab$  determined from the  $4\pi M(T)$ curves. Solid lines are guide for eyes.

The temperature dependence of the upper critical fields of C-doped MgB<sub>2</sub> for  $H \parallel c (H_{c2}^{c})$  and for  $H \parallel ab$   $(H_{c2}^{ab})$  defined as the onset of the diamagnetic response at  $4\pi M(H_{c2}) = 0$  were presented in Fig. 2. Temperature dependence of  $H_{c2}^{c}$  and  $H_{c2}^{ab}$  were found to be similar to those of pure MgB<sub>2</sub>;  $H_{c2}^{c}$  is almost linear and  $H_{c2}^{ab}$  presents a positive curvature near  $T_c$ , leading to a decrease of the anisotropy of  $H_{c2}(T)$  as the temperature approaches to  $T_c$ . While pure MgB<sub>2</sub> single crystals show a linear decrease in  $H_{c2}^{c}(T)$  near  $T_c$ , a close inspection reveals an upward curvature with C doping, which is consistent with the dirty-limit two-gap theory.

To analyze reversible magnetization data for  $H \parallel c$ , we used the Hao-Clem model [17]. Since the applied fields are comparable with  $H_{c2}^{c}(0)$ , only the Hao-Clem model, which considers not only the electromagnetic energy outside of the vortex cores, but also the free energy changes arising from the cores, can permits a reliable description of the reversible magnetization in the *entire* mixed state. However, for  $H \parallel ab$ , the simpler London model can be utilized because  $H_{c2}^{ab}(0)$  is much larger than the applied magnetic fields, therefore the contribution of

the core energy may not be as significant as it is for  $H \parallel c$ .

According to Kogan [18], the free energy of a uniaxial superconductor for which the anisotropy of the upper critical field,  $\gamma_H = H_{c2}^{\ ab} / H_{c2}^{\ c}$ , is different from the anisotropy of the penetration depth,  $\gamma_{\lambda} = \lambda_c / \lambda_{ab}$ , is given by

$$F = \frac{\phi_0 B \Theta_{\lambda}}{32\pi^2 \lambda_{ab}^2} \ln \left( \frac{2\sqrt{3}\gamma_H^{-2/3}\phi_0 \Theta_{\lambda}}{\xi^2 B (\Theta_{\lambda} + \Theta_H)^2} \right)$$
(1)

where  $\Theta_{\lambda,H}(\theta) = (\sqrt{\sin^2 \theta + \gamma_{\lambda,H}^2 \cos^2 \theta}) / \gamma_{\lambda,H}$ ,  $\phi_0$  is the flux quantum,  $\lambda_{ab}$  is the in-plane penetration depth, and  $\theta$  is the angle between the *c*-axis and the induction *B*. For  $H \parallel c$  and  $H \parallel ab$ , the free energy becomes

$$F(\theta=0) = \frac{\phi_0 B}{32\pi^2 \lambda_{ab}^2} \ln\left(\frac{\sqrt{3}\gamma_H^{-2/3}\phi_0}{2\xi^2 B}\right)$$

and

$$F(\theta = \frac{\pi}{2}) = \frac{\phi_0 B}{32\pi^2 \lambda_{ab} \lambda_c} \ln\left(\frac{2\sqrt{3}\gamma_H^{-2/3}\phi_0 \gamma_\lambda^{-1}}{\xi^2 B(\gamma_\lambda^{-1} + \gamma_H^{-1})^2}\right)$$
(2)

respectively. These results reduce to that of the original London model when the various anisotropies are equal to each other. With these equations, we calculated both the in-plane and the out-of-plane penetration depth.

From the relation  $M = -\partial F / \partial H$ , the magnetization can be calculated. For  $H \parallel ab$ , the magnetization gives

$$\frac{\partial M}{\partial \ln H} = \frac{\phi_0}{32\pi^2 \lambda_{ab} \lambda_c}$$
(3)

if it is assumed that the logarithmic term in the magnetization does not change drastically. When this equation is combined with  $\lambda_{ab}(T)$ ,  $\lambda_c(T)$  can be determined. Figure 3 shows  $\lambda_{ab}(T)$  and  $\lambda_c(T)$ 





Fig. 3. Temperature dependence of the in-plane  $[\lambda_{ab}(T)]$ and out-of-plane  $[\lambda_c(T)]$  penetration depth.  $\lambda_c(T)$  was calculated using the London model, and  $\lambda_{ab}(T)$  was calculated using the Hao-Clem model.  $\lambda_c$  and  $\lambda_{ab}$  are almost isotropic.

calculated from the Hao-Clem model and the London model, respectively. A notable feature is that  $\lambda_{ab}$  and  $\lambda_c$  are nearly isotropic in low temperature region and start to diverge as the temperature is increased, which implies that the anisotropy of  $\lambda$  is closed to ~1 at low temperatures and slowly increases with the temperature. This tendency agrees well with the theory with two gaps.

We compared the temperature dependence of  $\gamma_{\lambda}$ and  $\gamma_{H}$  in Fig. 4. The values of  $\gamma_{H}$  were deduced from the values of  $H_{c2}(T)$  shown in Fig. 2. Since  $\gamma_{\mu}(T)$  obtained from the reversible magnetization was in the limited temperature range, we added  $\gamma_{H}(T)$  obtained from the transport measurements. The temperature dependence of  $\gamma_H$  and  $\gamma_{\lambda}$  are observed to be similar to those of pure MgB<sub>2</sub>;  $\gamma_{H}$ decreases with temperature, and  $\gamma_{\lambda}$  is nearly temperature independent and converges to the value of  $\gamma_{H}$  near  $T_{c}$ . Two differences are observed in C-doped MgB<sub>2</sub>. First, the temperature dependence of  $\gamma_{H}$  is drastically reduced, which reflects that the anisotropic  $\sigma$  bands become more isotropic with C-doping. Second, the value of  $\gamma_{\lambda}$  decreases to ~1 over the temperature region measured. According to



Fig. 4. Temperature dependence of the anisotropy of  $\lambda(\gamma_{\lambda})$  and the anisotropy of  $H_{c2}$  that were obtained from reversible magnetization and transport measurements.

the theoretical calculation [11], impurity scattering could change the exact value of  $\gamma_{\lambda}$  and impurities in the  $\pi$  band drastically enhance  $\gamma_{\lambda}$ . Combined with the reduced temperature dependence of  $\gamma_{H}$ , this result provides an experimental support for relative increase of impurity scattering in the  $\sigma$  bands than in the  $\pi$  bands due to C-doping.

# 4. Summary

We have investigated the anisotropies of the penetration depth and the upper critical field of 10% C-doped MgB<sub>2</sub> single crystals, obtained from the reversible magnetization for magnetic fields applied both perpendicular and parallel to the c-axis of the crystals. The reversible magnetization was analyzed using the Hao-Clem model for  $H \parallel c$  and using the modified London model for  $H \parallel ab$ . The anisotropy of the upper critical field decreases with temperature while the anisotropy of the penetration depth remains nearly constant with temperature and two anisotropies converge at a value of 1 near  $T_c$ . The reduced temperature dependence of  $\gamma_H$  and the smaller value of  $\gamma_{\lambda}$ , compared to those in pure MgB<sub>2</sub>, indicate that C substitution enhances

impurity scattering mainly in the  $\sigma$  bands over the  $\pi$  bands. The temperature dependence of the anisotropies agrees well with the theoretical predictions with impurity scattering.

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